Anomalous Curie response of impurities in quantum-critical spin-1/2 Heisenberg antiferromagnets

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We consider a magnetic impurity in two different $S = 1/2$ Heisenberg bilayer antiferromagnets at their respective critical inter-layer couplings separating Néel and disordered ground states. We calculate the impurity susceptibility using a quantum Monte Carlo method. With intra-layer couplings in only one of the layers (Kondo lattice), we observe an anomalous Curie constant C^*, as predicted on the basis of field-theoretical work [S. Sachdev et al., Science **286**, 2479 (1999)]. The value $C^* = 0.262 \pm 0.002$ is larger than the normal Curie constant $C = S(S+1)/3$. Our low-temperature results for a symmetric bilayer are consistent with a universal C^*.

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Investigating effects of various types of impurities is a promising strategy for studying the electronic structure of strongly correlated systems [1, 2]. In very small concentrations, detectable changes in response functions cuaused by the impurities probe the inherent bulk properties of the host material and can give importants insights complementing information derived from direct studies of the bulk. This strategy was recently examined in a field-theoretical study [3, 4] of a nearly quantum-critical two-dimensional (2D) antiferromagnetic host system with a single localized spin-S impurity. Detailed predictions for the magnetic response of the impurity were made in the different finite-temperature scaling regimes associated with the quantum phase transition occuring in the host (as a function of some coupling constant g); from a quantum disordered spin-gapped paramagnet ($g > g_c$) to a gapless antiferromagnet ($g < g_c$). The $T \to 0$ asymptotic impurity susceptibility—defined as the difference between the susceptibilities of systems with and without impurity—was predicted to have a Curie form, $\chi^z_{\rm imp} \to C/T$, with the Curie constant C taking different values depending on the coupling [3];

$$C = S^2/3, \qquad g < g_c, \tag{1}$$

$$C = C^* = \tilde{S}(\tilde{S}+1)/3, \quad \tilde{S} \neq S, \qquad g = g_c, \tag{2}$$

$$C = S(S+1)/3, \qquad g > g_c, \tag{3}$$

where we have set $\hbar = k_B = 1$. The classical-like response for $g < g_c$, which can be understood as being due to the impurity spin aligning with a large cluster of correlated spins (exponentially divergent, thus with essentially classical dynamics as $T \to 0$), was confirmed in a recent numerical study [5], however only up to a logarithmic correction [formally resulting in a log-divergent $C(T)$]. The log-correction was subsequently derived quantitatively in a different field theoretical formulation [4] (and qualitatively also using spin wave theory [6]). It is given in terms of known ground-state constants of the host and is in complete agreement with the numerical results [7]. In the paramagnetic phase the host response is exponentially small due to the spin gap, and the usual Curie prefactor $S(S+1)/3$ is due solely to the localized impurity moment.

The most remarkable prediction of Ref. [3] is the anomalous Curie constant (2) in the quantum critical regime, i.e., for $T < \rho_s, \Delta$, where ρ_s is the spin stiffness in the Néel phase and the spin gap in the paramagnetic phase [8] (both of which vanish continuously at g_c). It was suggested that $S^2/3 < C^* < S(S+1)/3$ and that this could be interpreted in terms of a fractional impurity spin; $\tilde{S} \neq S$. However, the anomalous Curie constant was challenged by Sushkov [9], on the basis of a Green's function theory giving $\tilde{S} = S$ also in the quantum critical regime [10]. The only apparent way to settle this issue is by explicit unbiased numerical computation of the impurity susceptibility of a quantum-critical system. To our knowledge, the only attempt so far is by Troyer, who carried out a quantum Monte Carlo (QMC) study of the $S = 1/2$ Heisenberg bilayer with a vacancy (effectively corresponding to an $S = 1/2$ impurity) [11]. This calculation yelded $\tilde{S} = S$ within statistical errors of a few percent, and thus, based on this study, the anomaly is either not present or is very small.

In this Letter we present results of large-scale numerical efforts to resolve the controversy over the existence of an effectively fractional impurity spin at the quantum-critical point. We also obtain further insights into the temperature dependence of the impurity susceptibility (corrections to the Curie form). We will present evidence of an anomalous Curie constant in the case of an $S = 1/2$ impurity. Our result is $C^* = 0.262 \pm 0.002$, which falls outside the range $S^2/3 < C^* < S(S+1)/3$. It should be noted, however, that the ϵ-expansion presented for C^* in Ref. [3] was not evaluated explicitly—the actual value was only conjectured to fall within the above range. Thus there is no contradiction. For an $S = 1$ impurity we cannot detect an anomaly within statistical errors, indicating that C^* approaches $S(S+1)/3$ with increasing S.

We use an efficient stochastic series expansion (SSE) QMC technique [14] to compute the susceptibility of a single static $S = 1/2$ or $S = 1$ impurity in a quantum critical host. We consider two different host systems—a bilayer with intra- and inter-plane couplings J and $J_\perp$, respectively, and an "incomplete bilayer" in which the J-coupling is present only in one of the layers (a Kondo lattice [12]). The lattices and the ways in which we introduce the impurities in them are illustrated in Fig. 1. The Heisenberg hamiltonian for both host systems can be written as

$$\begin{aligned} H = \; & J \sum_{\langle i,j \rangle} \mathbf{S}_{1,i} \cdot \mathbf{S}_{1,j} + \lambda J \sum_{\langle i,j \rangle} \mathbf{S}_{2,i} \cdot \mathbf{S}_{2,j} \\ & + \; J_\perp \sum_i \mathbf{S}_{1,i} \cdot \mathbf{S}_{2,i}, \end{aligned} \tag{4}$$

where $\langle i,j \rangle$ denotes a pair of nearest neighbors on a periodic square lattice with $L \times L \times 2$ sites and $\mathbf{S}_{a,i}$ is the usual spin-$\frac{1}{2}$ operator at site i on layer $a = 1, 2$. We define the tuning parameter as $g = J_\perp/J$, and $J, J_\perp > 0$. The symmetric and incomplete bilayers correspond to $\lambda = 1$ and $\lambda = 0$, respectively. The quantum-critical points for these models have recently been extracted to high precision; $g_c = 2.5220(3)$ and $g_c = 1.3888(1)$ for the full and icomple bilayers, respectively [13]. The ground state is Néel ordered for $g < g_c$ and disordered (spin-gapped) for $g > g_c$. As shown in Fig. 1, by removing a single spin from the incomplete (a) and symmetric (b) bilayers we effectively introduce $S = 1/2$ impurities, as the remaining spin in the opposite layer is unpaired. An $S = 1$ impurity (c) is obtained by making the five bonds to a given spin ferromagnetic (we here keep the magnitudes of these interactions unchanged).

Our investigations proceed much in the same way as Ref. [11], but we have pushed the simulations to higher precision, larger lattices, and lower temperatures than previously. The calculations are very demanding due to the fact that the impurity susceptibility is the difference between two extensive quantities; the total susceptibilities with and without impurity. They are defined as

$$\chi_m^z = \frac{J}{T} \left(\sum_{i=1}^{N_m} S_i^z \right)^2, \tag{5}$$

with $N_0 = 2L^2$ when there is no impurity and $N_1 = 2L^2 - 1$ or $N_1 = 2L^2$ for the $S = 1/2$ and $S = 1$ impurities, respectively. The impurity susceptibility $\chi_{\rm imp} = \chi_1 - \chi_0$. Very large lattices are required in order to eliminate finite-size effects at low temperatures—we here report results for up to $256 \times 256 \times 2$ spins. We achieved relative statistical errors for χ_0 and χ_1 as small as $\approx 10^{-6}$. The use of improved estimators [15] is crucial, but still very long simulations are required. The calculations reported here required approximately 5×10^5 Pentium III ($\approx$ 1GHz) CPU hours.

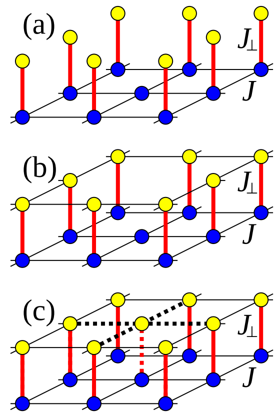

FIG. 1: (Color online) Spin impurity models. The circles represent $S = 1/2$ spins with nearest-neighbor interactions J (in-plane) and $J_\perp$ (inter-plane). A vacancy in the incomplete bilayer (a) or full bilayer (b) host constitutes, effetively, an $S = 1/2$ impurity. In (c), changing the signs ($J \to -J$ and $J_\perp \to -J_\perp$) of the interactions (the five dotted bonds) of one of the spins results in an $S = 1$ impurity.

Finite-size effects are investigated by considering systems, at their quantum-critical couplings, of increasing size $L = 4, 8, 16, \ldots$ at each temperature T, up to L sufficiently large for ramaining finite-zise corrections to be negligible. Finally, size-converged results are extrapolated to zero temperature to obtain the quantum-critical Curie constant defined in Eq. (2);

$$C^* = \lim_{T \to 0} T\chi_{\rm imp}^z. \tag{6}$$

The Curie form can be expected to apply strictly only in the limit $T \to 0$, and in practice we have to analyze corrections to extract C^*. In Ref. [11], finite-size scaled data for $T\chi_{\rm imp}^z$ were linearly extrapolated to zero temperature. We have also found an asymptotic linear correction in all cases studied, but we disagree with Ref. [11] in regards to the temperature at which the linear form is valid. In the case of a vacancy in the incomplete bilayer, our results extrapolate clearly to a value for C^* a few percent larger than $S(S + 1)/3$, thus confirming an anomalous Curie constant (falling outside the conjectured range, however). For the symmetric bilayer, which is the host system previously studied by Troyer [11], the linearity of the correction to C^* is established only at the lowest temperatures we have reached, and we cannot reliably extrapolate to obtain a direct independent confirmation of a universal (for given S) [3] anomalous Curie constant from these results. However, the available low-T data points are consistent with the results for the incomplete bilayer. For an $S = 1$ impurity we also find a linear correction to the Curie form but the extrapolated C^* is very close to the normal value.

We now present the data underlying our conclusions summarized above. In Fig. 2(a) we show the temperature dependence of $T\chi_{\rm imp}^z$ for the incomplete bilayer, for

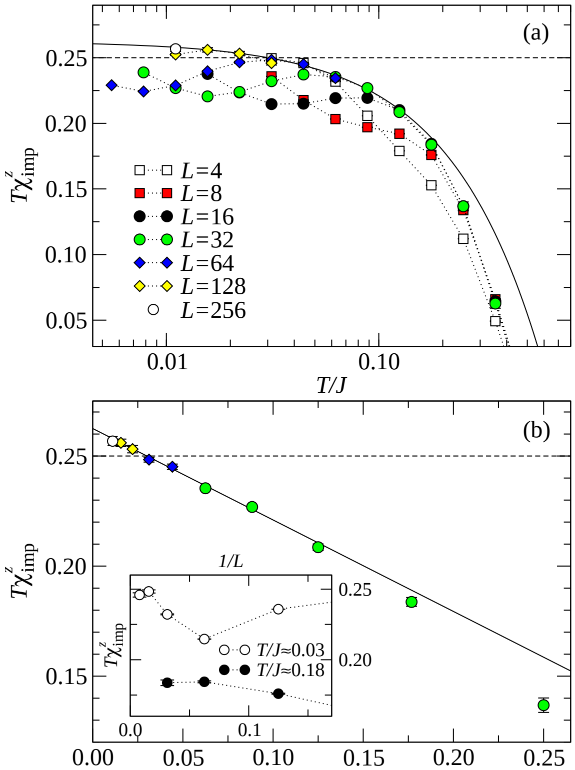

FIG. 2: (Color online) Impurity susceptibility multiplied by the temperature for a vacancy in the quantum-critical incomplete bilayer. (a) shows all our finite-size data on a log-lin scale and (b) shows size converged results on a lin-lin scale. Examples of finite-size effects are shown in the inset. The solid line in (b) and the curve in (a) show a linear in T fit to the low-T data.

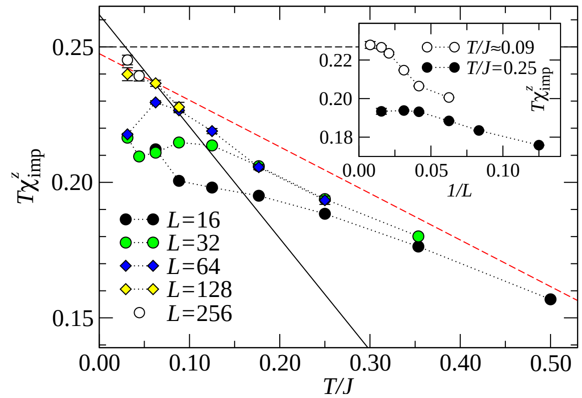

FIG. 3: (Color online) T times the impurity susceptibility for a vacancy in the quantum-critical symmetric bilayer. The solid line shows the linear fit of Fig. 2(b). The dashed (red) line is the linear fit of Ref. [11]. The inset shows examples of the size dependence.

all the system sizes we have considered and using a log-lin scale. Note first that as $T/J \to \infty$ the spins become independent moments exhibiting normal Curie behavior, and thus $T\chi^z_{\rm imp} \to -1/4$, due to there being one less spin in the system with a vacancy than in the intact system. We here focus on lower T. In the limit $T \to 0$, we have to obtain $T\chi^z_{\rm imp} \to +1/4$ for any L (seen in the figure only for $L = 4$), due to the $S = 1/2$ ground state of the even-L systems with a vacancy. The temperature at which this can be observed is expected to scale as $1/L$, reflecting the low-energy level spacing of a quantum-critical system with dynamic exponent $z = 1$ [8]. Interestingly, for the larger lattices the approach to the limiting $T = 0$ value is preceded by a miniumum and a maximum. The finite-size behavior for a fixed low T is thus also non-monotonic, as shown in the inset of Fig. 1(b). We consider $T\chi^z_{\rm imp}$ size-converged when results for system sizes L and $L/2$ agree to within statistical errors. We have checked this carefully for small systems and do not see any indications of non-monotonicities beyond a single minimum. Using this criterion we have size-converged data for temperatures down to $T/J = 1/64$.

At $T/J = 1/(64\sqrt{2})$, the infinite-size result is probably marginally higher than our $L = 256$ result. In Fig. 2(b) we show the size converged results [and the almost converged $T/J = 1/(64\sqrt{2})$ data] on a lin-lin scale. A linear T dependence can be seen below $T/J \approx 0.12$ and an extrapolation gives $C^* = 0.262(2)$. Note that even without an extrapolation it is clear that C^* exceeds the normal Curie constant $C = 1/4$ because the last three data points in Fig. 1(b) are all above this value.

To check for potential systematic errors, we have carried out extensive tests of our SSE codes, comparing results based on very long runs for 4×4 systems with exact diagonalization data and using two independently written programs with different random number generators for large lattices. Recent calculations using a completely different method have also confirmed the absence of systematic errors in SSE calculations [16]. Finally, we have also verified that small changes in the coupling g, of the order of the statistical errors of g_c [13], do not appreciably change $\chi_{\rm imp}$ in the range of tempertures studied here. We therefore consider our conclusion of an anomalous Curie constant to be beyond reasonable doubt.

In Fig. 3 we show data for the vacancy in a complete bilayer; the same system that was previously studied by Troyer [11]. The computational effort involved in simulating this system (for given T, L) is roughly twice that for the incomplete bilayer, because of the higher critical coupling and larger number of interactions. In addition, the size-convergence is slower. We have therefore not been able to go to as low temperatures as in the preceding case. We also find much larger corrections to the linear low-T behavior, and as a consequence we cannot carry out a reliable $T \to 0$ extrapolation. However, as shown in Fig. 3, the line fit to the low-T results for the incomplete bilayer in Fig. 2 describes reasonably well also the low-T results for the symmetric bilayer. The $L = 256$

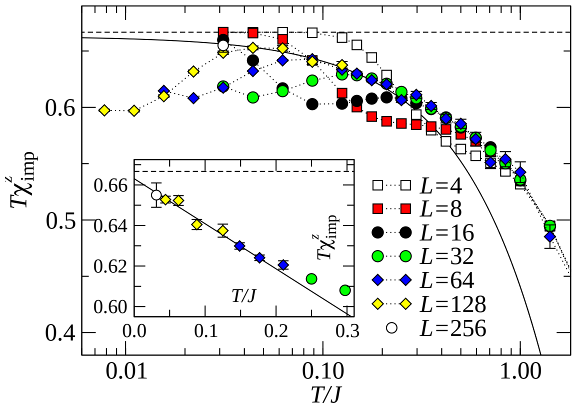

FIG. 4: (Color online) Impurity susceptibility of an $S = 1$ vacancy in the quantum-critical symmetric bilayer. The inset shows size converged results and a linear fit to the low-T data.

result at the lowest temperature is likely not completely size converged. The $L = 256$ point at the next-lowest T is also below the line, but the deviation is only two error bars and also here there may be some remaining finite-size effects. Moreover, the slope is most likely not universal and should thus depend on the low-energy parameters of the bulk system. The line shown may therefor not be the correct one for this system although C^* should be universal (for given S) [3, 4]. Thus we regard these symmetric bilayer results compatible with the anomalous C^* extracted for the incomplete bilayer.

In his study, Troyer fitted a line to QMC data in the range $0.1 \leq T/J \leq 0.4$ [11], resulting in the dashed line reproduced in Fig. 3—the $T = 0$ intercept is consistent with 1/4 within statistical errors. However, the line deviates considerably from our size-converged results for $T/J > 0.1$. The discrepancy may be due to a different size-convergence procedure; an $1/L$ extrapolation was mentioned in Ref. [11], whereas we have used a criterion of size-independence. The latter procedure should be more reliable because an asymptotically exponential convergence is expected at finite temperature. An $1/L$ extrapolation can lead to a too high value when fitting only to a range of points for which an almost linear in $1/L$ behavior is observed, as can be seen in the inset of Fig. 3 for $T/J = 0.25$.

We now turn to our results for an $S = 1$ impurity, which we have realized in the symmetric bilayer as shown in Fig. 1(c). As shown in Fig. 4, the finite-size behavior of the impurity susceptibility is similar to the $S = 1/2$ impurities. For $L = 4, 8$, and 16 the asymptotic low T behavior $T\chi^z_{\rm imp}(T \to 0) = S(S+1)/3 = 2/3$ is clearly seen. Size converged low-T results are shown in the inset. A linear behavior sets in at $T/J \approx 0.15$. Here the extrapolated $C^* = 0.663(2)$ is slightly below the normal value 2/3, but the difference is too small to definitely conclude that this is the case. The results do show that the anomaly is smaller than for $S = 1/2$. The theory does not predict how the fractional spin evolves as a function of S [3, 4].

In summary, we have presented evidence from unbiased numerical computations of an anomalous Curie response of an $S = 1/2$ impurity spin in a 2D quantum-critical antiferromagnet. The anomalous Curie constant $C^* = 0.262(2)$ is only $\approx 5\%$ larger than the normal $C = 1/4$. For $S = 1$ we obtain C^* marginally below the normal value, 2/3, but better statistics is needed to confirm this.

It should be noted that the anomalous Curie constant cannot be strictly interpreted as due to a fractionalized impurity spin, because it is a finite-temperature quantity with contributions from many states with different total spin, even as $T \to 0$. Fractionalization does not occur in the ground state for finite L, but an interesting universal impurity-induced spatial structure has been found [17]. Recently impurity effects have been examined theoretically also in fractionalized spin liquid states [18, 19].

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